Automation of Radiochemical Analysis by Flow Injection Techniques

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Flow injection belongs to a family of analytical methods based on a sample injection into a flowing unsegmented stream which transports the analyte through a chemical modulator into the detector. Unlike chromatographic techniques, flow injection exploits chemical reactions to transform the analyte into the detectable species under well defined fluidic conditions. Flow injection found great success as a laboratory technique for automation of reagent based assays and as the means of enhance the performance of a variety of analytical detectors. Recent advances in flow injection methodology and instrumentation, introduction of stopped-flow techniques, and most recently, sequential injection techniques made flow injection a highly versatile automated solution handling platform with numerous successful applications in various areas of analytical chemistry.

Many of the basic characteristics of the flow injection methodology are very attractive for the automation of radiochemical methods. All solution handling operations are automated and fully contained, leading to minimized personnel exposure and elimination of the human bias. All sample manipulations are performed under highly reproducible and well defined fluidic and geometrical conditions, a very desired feature for radioactivity detection. Stopped-flow methodology can be used to increase the residence time of a sample in the detector cell and thus to overcome the fundamental limitation of the radioactivity detection in a flow mode. Flow injection separation methodology is well documented and can be adapted to radionuclide separations. Finally, flow injection methods feature significant reduction of secondary waste compared to batch analytical procedures.

Recently developed highly selective sorbents for radionuclide separations marketed by EIChrom Industries Inc. allow to perform radionuclide separations in a format that is highly suitable for automation by flow injection techniques. Integrated with commercially available radioactivity detection instrumentation, automated radionuclide separations can be used to develop flow injection-based radionuclide analyzers. Flow injection instrument can also serve as an automated separation workstation if interfaced with a fraction collector.

Despite the recognized potential, thus far there has been very little application of low injection to radionuclide separations and analysis. The ongoing research in our laboratories is aimed to correct this situation and is focused on the development of flow injection methodologies for radionuclide separations and analysis with the ultimate goal of creating an automated flow injection "tool kit" suitable for routine applications in radiochemical laboratories at the Hanford Site.

Recently we developed a rapid automated procedure for the determination of 90Sr in aged nuclear waste. The procedure is based on a sequential injection analysis system which rapidly separates 90Sr from interfering radionuclides on a minicolumn containing Sr-Resin=99. The strontium is nearly selectively retained on the column in high molarity nitric acid solutions, while 90Y and most of the interfering radionuclides are removed from the column with a few free column volumes of the nitric acid wash. The separated Sr is removed from the column with water and its radio activity is detected on-line using a flow through liquid scintillation detector. Sample radioactivity can be quantified from the peak areas giving linear calibration curves. The instrument can be operated in a stopped-flow regime to improve the sensitivity of the on-line radioactivity detection, while the system separates the next sample. The novel analytical methodology has been characterized in detail and its effectiveness has been demonstrated on the analysis of the aged tank waste samples from the Hanford Site. The advantages of the new technique included full automation, faster analysis, greater precision, reduced labor costs, and reduced waste generation.

Our most recent results demonstrate that flow injection can be successfully used in automation of substantially more difficult separations such as the separation of actinide elements. We developed an automated selective Pu-Am separation on TRU Resin=99 that requires on-the-column adjustments of the Pu oxidation state to enable its efficient separation from An (III, IV, VI). Precise control over fluidic conditions, use of the smaller size sorbent particles, possibility to change the experimental parameters from computer keyboard, while the progress of the separation is monitored in real time, allows to "fine tune" radionuclide separations and develop more efficient

analytical protocols. The experiments that are too tedious to be performed in a manual format can be easily done in flow format allowing to obtain new information leading to improved performance of the separation materials.

Reliable performance in the real world environment places strict requirements on the quality and reliability of flow injection instrumentation. Introduction of a new generation of flow injection instrumentation provides a hardware base suitable for technology transfer applications.

In general, the results obtained thus far allow us to safely state that flow injection emerges as a very promising tool for automation in radiochemistry and has a potential to improve the current state of the art of a wide range or radioanalytical methods.